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ABSTRACT

A universal solvent extraction (UNEX) process for the simultaneous separation of cesium, strontium, and the actinides from actual radioactive acidic tank waste was demonstrated at the Idaho National Engineering and Environmental Laboratory. The waste solution used in the countercurrent flowsheet demonstration was obtained from tank WM-185. The UNEX process uses a tertiary solvent containing 0.08 *M* chlorinated cobalt dicarbollide, 0.5% polyethylene glycol-400 (PEG-400), and 0.02 *M* diphenyl-N,N-dibutylcarbamoyl phosphine oxide (Ph_2Bu_2CMPO) in a diluent consisting of phenyltrifluoromethyl sulfone (FS-13). The countercurrent flowsheet demonstration was performed in a shielded cell facility using 24 stages of 2-cm diameter centrifugal contactors. Removal efficiencies of 99.4%, 99.995%, and 99.96% were obtained for ^{137}Cs, ^{90}Sr, and total alpha, respectively. This is sufficient to reduce the activities of ^{137}Cs, ^{90}Sr, and actinides in the WM-185 waste to below NRC Class A LLW requirements. Flooding and/or precipitate formation were not observed during testing. Significant amounts of the Zr (87%), Ba (>99%), Pb (98.8%), Fe (8%), Ca (10%), Mo (32%), and K (28%) were also removed from the feed with the universal solvent extraction flowsheet. ^{99}Tc, Al, Hg, and Na were essentially inextractable (<1% extracted).

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INTRODUCTION

Separation processes are being evaluated for the treatment of acidic high-activity tank waste at the Idaho National Engineering and Environmental Laboratory (INEEL) with the goal of minimizing the high-activity waste volume to be disposed in a deep geological repository. The Idaho Nuclear Technology and Engineering Center (INTEC), formerly known as the Idaho Chemical Processing Plant (ICPP), is the only facility storing high-activity waste at the INEEL. Nearly 5 million liters of aqueous acidic high-activity waste, known as sodium-bearing waste (SBW), are currently on inventory at the INTEC. This waste was derived primarily from solvent washing operations in the uranium recovery process and equipment decontamination activities. The INTEC is no longer recovering uranium; therefore, waste from this process is no longer being generated. However, waste generation from decontamination activities and daily plant operations is continuing.

SBW is stored in underground stainless steel tanks. A Notice of Noncompliance was filed in 1992 by the State of Idaho Department of Health and Welfare and the Region 10 EPA contending that the tanks did not meet secondary containment requirements set forth in Title 40, Part 265.193 of the Code of Federal Regulations. A recent agreement between the State of Idaho, Department of Energy, and United States Navy, known as the 1995 Settlement Agreement, establishes that the SBW must be out of the tanks by 2012. Separation processes are being evaluated as alternatives to remove the waste from the tanks by 2012, while at the same time, treat the waste for permanent disposal.

The INTEC has historically blended SBW with fuel reprocessing raffinates and solidified it in a fluidized bed calciner operated at 500 ^{o}C. However, all the fuel reprocessing raffinates were depleted by calcination in 1993, which eliminated the option of blending these two wastes. SBW cannot be calcined by itself because the high sodium content causes bed agglomeration in the calcination vessel. SBW can be blended with non-radioactive aluminum nitrate as another option for calcining the waste and removing it from the tanks by 2012. However, this option increases the calcine volume, and the resulting calcine would still require further treatment before final disposal. Additionally, proposed legislation and permitting requirements would require extensive modifications to the calcination facility prior to operation beyond April of 2000. Separation is also an option for treating INTEC high-activity calcined waste. Therefore, calcined SBW may eventually be treated by separating the radionuclides from the inert components of the waste.

Several separation technologies have been demonstrated at the INEEL using actual SBW. The TRUEX process has been demonstrated to efficiently remove the transuranic (TRU) elements from the waste in a 2-cm centrifugal contactor pilot-plant.[1,2] Likewise, strontium removal has been demonstrated in the 2-cm centrifugal contactor pilot-plant using the SREX process.[3,4] Cesium removal has been demonstrated in small ion exchange columns (1 - 1.5 cm^3) loaded with either potassium hexacyanoferrate, crystalline silicotitanates, or ammonium molybdophosphate.[5,6] Finally, collaborative testing efforts between Russian scientists from the V. G. Khlopin Radium Institute in St. Petersburg, Russia, have resulted in the successful demonstration of the chlorinated cobalt dicarbollide process with and without polyethylene glycol (PEG) for the removal of cesium and strontium (with PEG) from INTEC SBW.[7,8] These collaborative tests have also resulted in the successful demonstration of a phosphine oxide process for removing TRU's from SBW.[8]

The use of a single process to remove the desired radionuclides, as opposed to a combination of different unit operations that remove these same radionuclides, evolved from the previous collaborative work with the Russian scientists. The possibility of using a universal solvent containing chlorinated cobalt dicarbollide with PEG to remove cesium and strontium, and a carbamoylmethyl phosphine oxide derivative to remove the TRU's was discussed early in FY-95. A proposal to investigate such a solvent was submitted to and accepted by the Department of Energy Office of Science and Technology Efficient Separations and Processing Cross-Cutting Program. A process based on a universal solvent may provide a more simple and cost effective method for waste treatment than a method that utilizes two or three separate processes. Batch contact testing of the universal solvent was performed in 1997 using actual INTEC SBW, a countercurrent flowsheet test using 26 stages of 3.3-cm diameter centrifugal contactors and simulated tank waste was performed in 1997, and a countercurrent flowsheet test using 24 stages of 2.0-cm diameter centrifugal contactors and actual tank waste was performed in 1998, all with very positive results.[9-11] The flowsheet demonstration performed with actual waste in 1998 resulted in removal efficiencies of 99.9% and 99.99% for Cs and Sr, respectively. However, the removal efficiency obtained for the actinides was only 96%. This low removal efficiency was attributed to loading of the Ph_2Bu_2CMPO extractant in the universal solvent with metals such as Zr, Fe, and Mo. Based on these results, further testing was performed at the Khlopin Radium Institute and a modified flowsheet was developed to suppress the extraction of metals. This flowsheet was demonstrated using actual INTEC SBW in a centrifugal contactor pilot plant located in a shielded cell facility.

This document reports the results of the Universal Extraction (UNEX) process flowsheet demonstration with SBW. Distribution coefficients and removal efficiencies of the actinides, ^{137}Cs, ^{90}Sr, and some of the non-radioactive elements are reported. Observations related to flooding and precipitate formation are also reported.

PURPOSE AND SCOPE

The purpose of this study was to demonstrate a UNEX flowsheet developed by the Khlopin Radium Institute and the INEEL for the separation of Cs, Sr, and the actinides from INTEC SBW. The flowsheet for the treatment of INTEC SBW was evaluated by performing countercurrent flowsheet tests using 24 stages of 2.0-cm diameter centrifugal contactors.

The countercurrent flowsheet demonstration was performed using actual SBW from tank WM-185. The ability to reduce the activity of Cs, Sr, and the actinides to below NRC Class A LLW limits and the behavior of the metals with the flowsheet was evaluated. Ultimately, the activity of Cs, Sr, and the actinides in the final waste form for the low-activity waste (e.g. grout) are desired to be below the NRC Class A LLW limits. The effect of volume change on activities resulting from making of the LAW waste form must be evaluated in order to determine if Class A limits are met.

To support the flowsheet testing, batch contact tests using the universal solvent and WM-185 tank waste were also performed.

EQUIPMENT DESCRIPTION

The flowsheet demonstration was performed using 2-cm diameter centrifugal contactors installed in the CPP-684 Remote Analytical Laboratory (RAL) shielded hot cell. The centrifugal contactors, as shown in Figure 1, consist of 24 stages of 2-cm diameter centrifugal contactors, feed and receiving vessels, feed pumps, and an air purge system for the contactor bearings. The aqueous and organic feed pumps and feed vessels were located inside the shielded cell. The remaining feed pumps and feed vessels were located outside the cell. All of the feed pump controllers were located outside the cell. Non-radioactive solutions used for the flowsheet testing were pumped to the centrifugal contactors through penetrations in the cell wall.

The centrifugal contactors were designed and fabricated by Argonne National Laboratory. The centrifugal contactors were designed specifically for operation of the TRUEX process with INTEC SBW. The contactors were modified at the INEEL for remote installation and operation in the RAL hot cell. Specifically, a modified support structure was fabricated for the contactors. This support structure is portable to allow the contactors to be moved out of the way when not in operation, contains leveling screws to adjust for unevenness in the cell floor, and can be disassembled into three sections. It was necessary to design the support structure for disassembly and re-assembly so that the structure would fit through the 12-in. by 22-in. glove box access port into the cell and be assembled remotely. The centrifugal contactors were installed through the access port in groups of four and assembled on the support structure remotely. Lifting bails were installed on each contactor to facilitate remote replacement or inspection of any motor/rotor assemblies. A description of the centrifugal contactors is provided in Table 1.

Solution was fed to the contactors using valveless metering pumps. Surge lines, consisting of 4-inch sections of 1-inch stainless steel tubing, were placed on the outlet of the pumps to dampen surges in solution flow. Because of the difficulty associated with remote installation, surge lines were not installed for the aqueous and organic feed pumps located in cell. Flowrates were adjusted by controlling the pump speed using a ten-turn potentiometer or by manually adjusting the piston stroke length.

Clear, flexible Teflon® or Teflon® lined Tygon® tubing was used for inlet and outlet connections to the feed and receiving vessels. The feed lines were 1/8-in. o.d. tubing and the product lines were 3/8-in. o.d. tubing.

The 2-cm centrifugal contactors do not have provisions for sampling the aqueous and organic solutions exiting individual stages during operation. The aqueous raffinate, strip product, wash effluent, and solvent recycle streams were sampled by routing the solution draining to a receiving vessel into a sample bottle during the actual flowsheet test. Individual stage samples were taken by draining the contactor stages after shutdown.

Table 1. Description of the 2-cm centrifugal contactors.

Size	2-cm rotor diameter
Motor	115 Volt, 60 Hz Bodine Model 710
RPM	3,600 rpm (not adjustable)
Material of construction	304L stainless steel
Inlet and outlet ports	3/8-in. o.d. tubing
Configuration	Single stage units which can be configured as desired. Stages are connected using U- tubes.

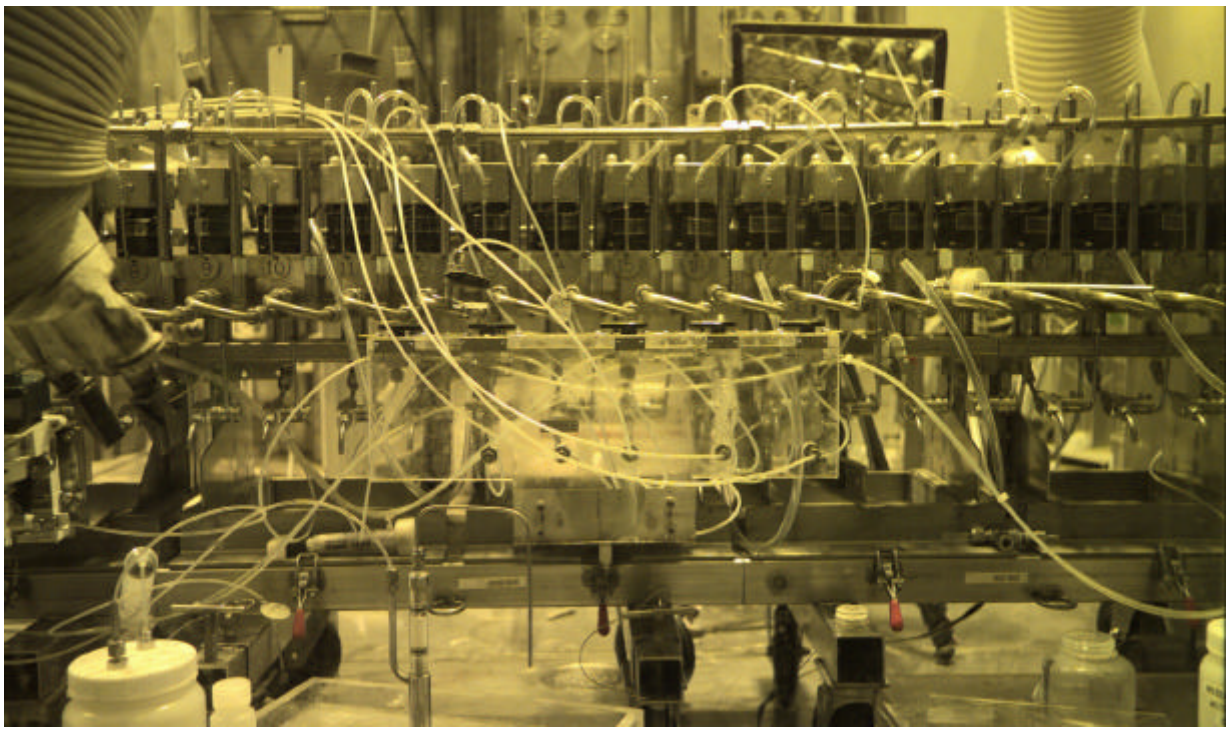

Figure 1. 2-cm diameter centrifugal contactors installed in the Remote Analytical Laboratory shielded cell.

An air purge system was connected to the contactor bearing housings. Purge air was required in order to protect the motor body, shaft, and bearings from corrosive process fumes. Air to the bearing housings passed through a rotameter. The air flowrate through the rotameter could be adjusted from zero to five scfh. Air from the rotameter was split to feed each of the 24 contactors. The diameter of the air manifold was large (1/2-in. o.d. tubing), while the diameter of the tubes leading from the manifold to the contactors was small (1/8-in. o.d. tubing), resulting in the airflow to each contactor being approximately equivalent. The offgas from the bearing purge system was vented to the cell.

METHODOLOGY/EXPERIMENTAL PROCEDURE

WM-185 Tank Waste

Approximately 0.9 liters of actual INTEC SBW, obtained from tank WM-185 in 1999 was used as feed solution for the flowsheet testing. The feed solution was first filtered through a 0.45-micron filter. Currently, approximately 5.0 million liters of SBW are stored in six tanks. The composition in each tank varies; however, the composition of the solution in tank WM-185 is representative of the solution in all the tanks. The chemical composition of the average composition of the current SBW inventory is shown in Table 2. For the flowsheet demonstration with WM-185 tank waste, 90 mL of 5.2 *M* HF was added to 810 mL of feed. The feed is diluted to reduce the concentration of metals, thus decreasing the loading of the actinide extractant in the solvent. Fluoride is added to the feed to complex Zr and Fe, thus minimizing the extraction of Zr and Fe. The composition of the adjusted WM-185 feed is shown in Table 2.

Solvent

The UNEX process solvent used in all testing is a "universal extraction mixture" which was developed to remove all of the radionuclides of concern, both fission product and actinides, in a single solvent extraction unit operation. The solvent is designed to be the heavy phase, i.e., it has a specific gravity greater than the aqueous solutions. The solvent composition tested was 0.08 *M* chlorinated cobalt dicarbollide, 0.5% polyethylene glycol-400 (PEG-400), and 0.02 *M* diphenyl-N,N-dibutylcarbamoyl phosphine oxide (Ph_2Bu_2CMPO) in a diluent consisting of phenyltrifluoromethyl sulfone (FS-13).

The solvent used for the batch contact tests and for the countercurrent flowsheet demonstration was freshly prepared several days prior to testing. Approximately 1,000 mL of solvent was available for the flowsheet test. As a result, it was necessary to recycle the solvent continuously during testing. Based on the solvent feed rate of 8 mL/min and 315 minutes of solvent feed, the solvent was recycled a total of 2.5 times. Of the total 315 minutes of solvent feed, 255 minutes was with WM-185 feed. This corresponds to the solvent being recycled 2.0 times while operating with WM-185 feed. Approximately 55 mL of the freshly prepared solvent was retained and used in the batch contact tests with tank waste. Consequently, all of the tests discussed initially used fresh, unused solvent.

Table 2. Adjusted WM-185 tank waste and average SBW tank waste compositions.

Component	Adjusted WM-185	Average SBW	Component	Adjusted WM-185	Average SBW
Acid (*M*)	1.45	1.59	Na (*M*)	1.06	1.9
Al (*M*)	0.51	0.64	NO_3 (*M*)	---	5.07
B (*M*)	---	0.018	Zr (*M*)	0.010	0.002
Ba (*M*)	4.9E-05	6.1E-05	Alpha (nCi/g)	525	369
Ca (*M*)	0.050	0.054	^{241}Am (nCi/g)	50	90
Cr (*M*)	---	0.003	^{134}Cs (Ci/m^3)	---	1.1
F (*M*)	---	0.070	^{137}Cs (Ci/m^3)	88	41
Fe (*M*)	0.021	0.022	^{238}Pu (nCi/g)	435	245
Pb (*M*)	0.0018	0.0012	^{239}Pu (nCi/g)	53	35
Hg (*M*)	0.0037	0.0011	^{99}Tc (Ci/m^3)	0.031	---
Mo (*M*)	0.0002	0.0006	^{90}Sr (Ci/m^3)	75	38
K (*M*)	0.14	0.206	U (g/L)	---	0.12

Analytical

Samples from testing were analyzed for total alpha, ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{241}Am, ^{99}Tc, ^{154}Eu, Al, Ba, Ca, Ce, Fe, Hg, K, Mo, Nd, Na, Pb, and Zr. Alpha spectroscopy was used for Am and Pu analyses, and gamma spectroscopy was used for Cs and Eu analyses. Inductively coupled plasma emission spectroscopy (ICP-ES) was used for Al, Ca, Ce, Nd, Pb, Mo, Zr, Ba, and Fe analyses. Atomic absorption spectrophotometry (AAS) was used for Na and K analyses, and atomic fluorescence spectroscopy was used for Hg analyses. Strontium analyses were performed by separation with the classical fuming nitric acid technique. Strontium was then precipitated onto a glass filter and analyzed by a glass flow proportional counter. Total alpha analyses were performed by evaporating a sample aliquot on a 52-mm stainless steel counting planchet. Samples were then counted by gas flow proportional counting.

Organic samples could not be directly analyzed for ^{90}Sr, ^{238}Pu, ^{239}Pu, ^{241}Am, ^{99}Tc, Al, Ba, Ca, Ce, F, Fe, Hg, K, Mo, Nd, Na, Pb, and Zr. Therefore, each organic dilution from the SBW tests was contacted with a solution containing 1.5 *M* guanidine carbonate and 25 g/L diethylenetriamine pentaacetic acid (DTPA) at an organic to aqueous phase ratio of 0.2. The resulting aqueous phase was used for analysis of the organic samples. This method of indirect analysis assumes that all of the analytes are stripped from the organic phase by the aqueous solution.

Batch Contact Flowsheet Testing With WM-185 Tank Waste

Prior to performing the UNEX flowsheet demonstration, a batch contact flowsheet was performed with actual WM-185 waste solution. The purpose of this testing was to determine distribution coefficients of ^{137}Cs, ^{90}Sr, ^{85}Sr, and ^{154}Eu for the extraction and stripping contacts, and to observe the batch contacts to determine if physical problems such as precipitate formation are present. The batch contact flowsheet test is summarized in Figure 2.

Feed for the batch contact flowsheet test was prepared by adding 30.6 mL of scrub solution (0.3 *M* citric acid in 0.05 *M* HNO_3) and 15.4 mL of 5.2 *M* HF to 154 mL of WM-185 tank waste. The feed solution was also spiked with ^{85}Sr. This feed solution was contacted with the universal solvent at an organic to aqueous phase ratio (O/A) of 1 (contact E1). The resulting organic solution from contact E1 was again contacted with feed solution at an O/A of 1 (contact E2). This process was repeated two more times (E3 and E4). The resulting organic from contact E4 was then scrubbed once with 0.3 *M* citric acid in 0.05 *M* HNO_3 at an O/A of 1 (Sc1), followed by two strip contacts (St1 and St2) with 1 *M* guanidine carbonate and 10 g/L DTPA at an O/A of 1. A separate extraction contact (contact E5) was performed using un-adjusted WM-185 and the universal extractant at an O/A of 1. This contact was performed as a comparison to determine the effect of adjusting the feed solution on distribution coefficients. A third strip contact (St3) was also performed using the organic from St1 and a 1 *M* guanidine carbonate, 20 g/L DTPA strip solution to determine the effect of increasing the DTPA concentration on the actinide strip distribution coefficient. All contacts were made by placing the aqueous and organic solutions into a poly bottle, shaking the bottle with a manipulator for three minutes, and separating the two phases in a separatory funnel. Observations regarding precipitation formation and phase disengagement time were recorded.

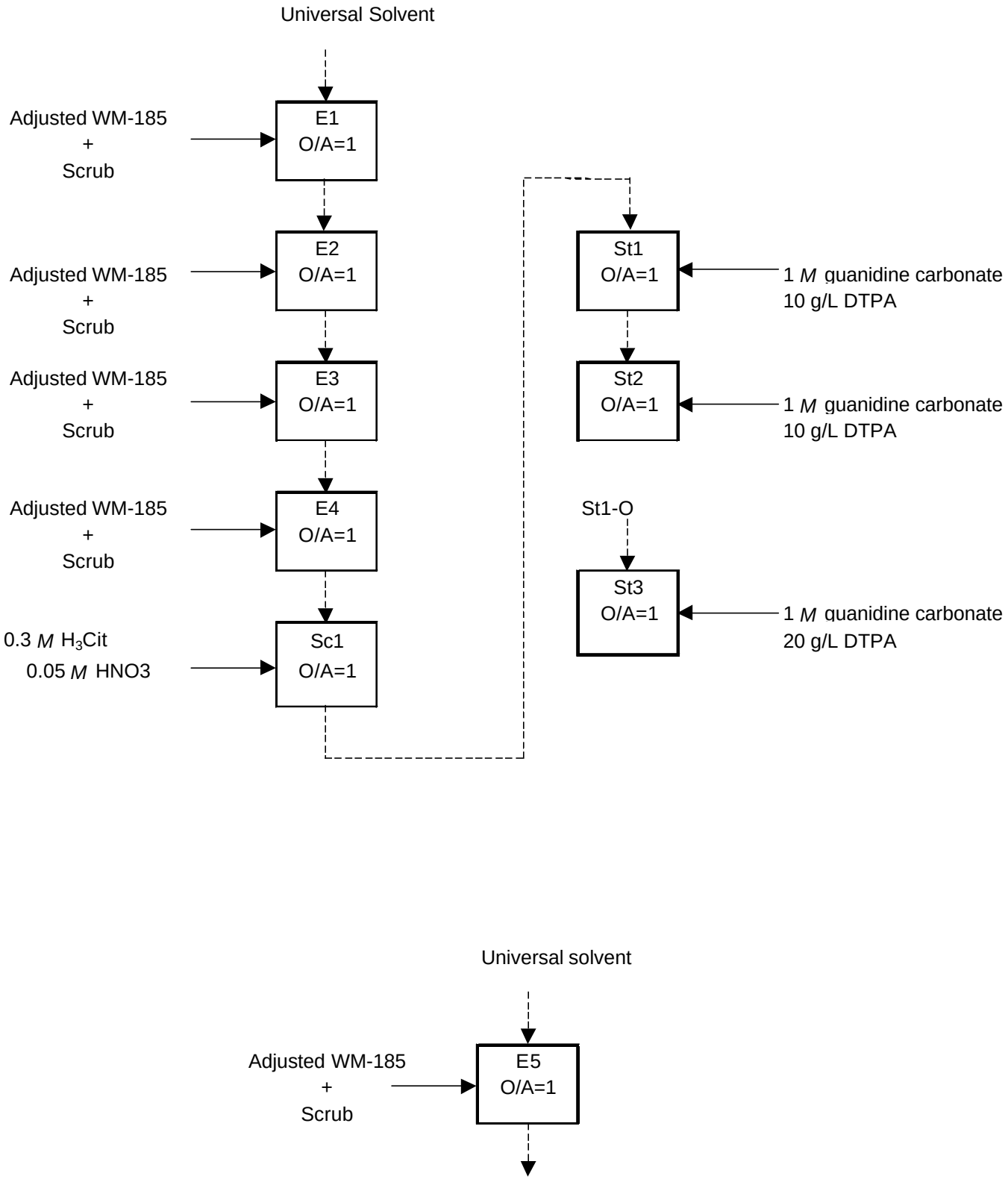

Figure 2. Batch contact flowsheet for testing with WM-185 tank waste.

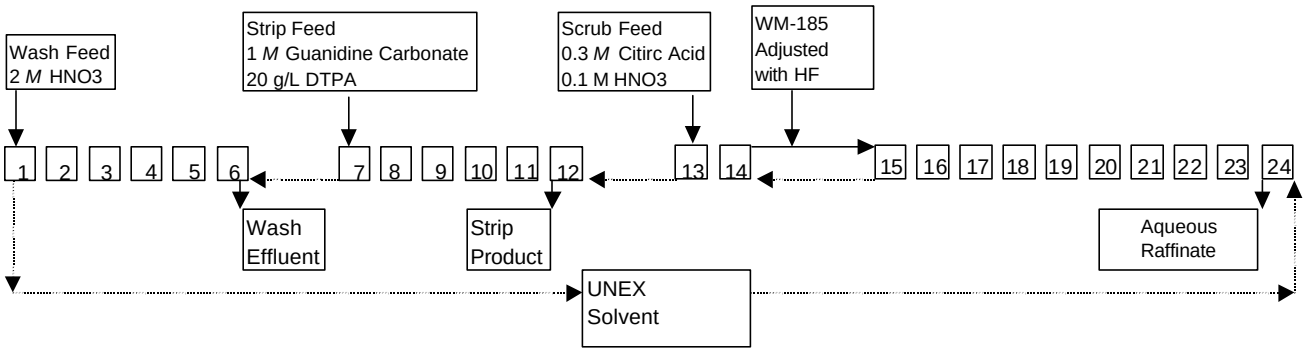

Figure 3. Flowsheet for testing of the UNEX process with WM-185 tank waste.

Flowsheet Demonstration With WM-185 Tank Waste

Based on the results of universal solvent development studies performed at the Khlopin Radium Institute and at the INEEL, a flowsheet was developed and recommended for countercurrent flowsheet testing in the 2-cm diameter centrifugal contactors using actual waste solution. This flowsheet, as shown in Figure 3, consists of ten stages of extraction at an organic to aqueous phase ratio (O/A) of 2.0, two stages of scrub (0.3 *M* citric acid in 0.1 *M* HNO_3) at an O/A of 8.0, six stages of strip (1 *M* guanidine carbonate, 20 g/L DTPA) at an O/A of 1.5, and six stages of solvent wash (2 *M* HNO_3) at an O/A of 1.5.

The flowsheet demonstration was performed as follows. Each of the centrifugal contactors were filled with 15 mL of process solution by pumping the appropriate solution into each stage through the overflow ports. One molar HNO_3 was used for the stages in the extraction section. The centrifugal contactor motors were then started at 3,600 rpm. Solvent flow was established. When solvent began exiting contactor stage 1, aqueous solution flows were established. One molar HNO_3 was used in place of the WM-185 feed for the startup. Approximately the first 50 mL of solvent to exit the contactors was collected separately in case it picked up contaminants from previous flowsheet testing. The process then continued to operate without recycle of the solvent. Twenty minutes after the start of the aqueous flows, WM-185 flow was established. Samples were taken from the raffinate 60 and 80 minutes after the start of WM-185 feed. At this time recycle of the solvent was initiated and the raffinate, strip product, wash effluent, and solvent effluents were sampled at intervals of 30 minutes. Final samples of all effluent streams were taken 255 minutes after the start of actual waste feed. The contactors were then shut down by simultaneously stopping the contactor motors and feed pumps. Each stage remains approximately at steady-state operating conditions with this type of shutdown. This allowed aqueous and organic samples to be taken from each stage and, therefore, distribution coefficients to be determined for any of the 24 stages.

RESULTS AND DISCUSSION

Batch Contact Flowsheet Testing With WM-185 Tank Waste

Results of the batch contact flowsheet test are summarized in Table 3. Extraction distribution coefficients ranged from 1.1 to 1.2 for ^{137}Cs, 1.3 to 2.2 for ^{90}Sr, 1.3 to 2.7 for ^{85}Sr, and 2.0 to 5.6 for ^{154}Eu (Am surrogate). The strip solution effectively stripped the Cs and Sr from the universal solvent. The Eu strip distribution coefficients of 0.44 and 0.99 were higher than desired for effective stripping. However, the Eu strip distribution coefficient decreased to < 0.05 when the DTPA concentration in the strip feed was increased from 10 g/L to 20 g/L.

In the E5 contact, which was the same as the E1 contact except unadjusted WM-185 feed was used, the Cs, Eu, and Sr distribution coefficients were significantly lower. This is as expected since the concentrations of nitric acid and metals are higher, and no fluoride has been added to suppress the extraction of Zr and Fe.

As a result of the batch contact testing, adjusted feed, identical to the adjusted feed for the batch contact testing, was used. Also, a DTPA concentration of 20g/L in the strip feed was used.

Table 3. Results of batch contact flowsheet testing with WM-185 tank waste.

Contact	D_{Cs-137}	D_{Eu-154}	D_{Sr-85}	D_{Sr-90}	Phase Disengagement time (min)	Observations
E1	1.2	2.0	2.7	2.2	1.75	Clear
E2	1.1	5.6	1.8	---	1.6	Clear
E3	1.1	3.8	1.6	---	2.7	Clear
E4	1.1	3.5	1.3	1.3	2.3	Clear
St1	0.18	0.99	<0.01	---	0.42	Clear
St2	0.28	0.44	---	---	0.33	Clear
St3	0.10	<0.05	---	---	---	---
E5	0.68	0.20	---	1.7	1.5	Clear

UNEX Flowsheet Demonstration with WM-185 Tank Waste

Contactor Operation

Actual solution flowrates for the flowsheet demonstration were calculated from feed tank depletion rates and are compared to the desired flowrates in Table 5. Desired and actual flowrates compare very well for each of the feed streams. Flooding was not observed in the effluent streams. Previous flowsheet testing resulted in flooding in the actinide strip section of the flowsheet.[11] Actinides were recycled back to the extraction section, thus reducing the removal efficiency. The total flowrate in the strip section was reduced from 16.1 mL/min to 12.9 mL/min for this testing to reduce the possibility of flooding. Precipitate formation was not observed in any of the samples taken during operation or in the contactors after shutdown.

The temperature in the RAL hot cell the morning of the test was 32 ºC. The extraction distribution coefficients for Cs, Sr, and the actinides decrease with increasing temperature. A temperature increase from 25 ºC to 36 ºC results in a 17%, 13%, and 56% decrease in the distribution coefficients for Cs, Sr, and Eu, respectively. To minimize the adverse effects of the high cell temperature, the solvent feed container was placed in small ice bath. Temperature of the solvent was not recorded but previous attempts to cool the solvent with this method resulted in a solvent temperature of 26 ºC to 30 ºC.

Table 4. Flowrates and O/A ratios for the flowsheet demonstration with WM-185 tank waste.

		Flowrate (mL/min)		O/A Ratio		Total Flow
Section	Phase	Desired	Actual	Desired	Actual	(mL/min)
All	Org.	8.0	7.9	---	---	---
Extraction	Aq.	4.0	4.0	2.0	2.0	11.9
Scrub	Aq.	1.0	0.9	8.0	8.8	8.8
Strip	Aq.	5.3	5.0	1.5	1.6	12.9
Wash	Aq.	5.3	5.0	1.5	1.6	12.9

Table 5. Percentages of radionuclides in the effluent streams for the flowsheet demonstration with WM-185 tank waste.

Effluent	^{137}Cs	^{90}Sr	Alpha	^{241}Am	^{238}Pu	^{239}Pu	^{154}Eu	^{99}Tc
Raffinate	0.57%	0.0052%	0.040%	0.0002%	0.006%	0.002%	0.42%	81.2%
Strip	100.6%	108.1%	100.4%	105.6%	96.9%	103.%	78.6%	<0.14
Wash	0.006%	0.0003%	0.001%	---	---	---	0.015%	---
Solvent	0.005%	0.005%	0.02%	0.2%	0.005%	0.0006%	0.075%	0.013%
Material Balance	101.1%	108.1%	100.4%	105.8%	97.0%	103.9%	79.1%	81.3%

Compositions at Shutdown

The percentages of ^{137}Cs, ^{90}Sr, total alpha, ^{241}Am, ^{238}Pu, ^{239}Pu, ^{154}Eu, ^{99}Tc, Al, Ba, Ca, Ce, Fe, Hg, K, Mo, Na, Nd, Pb, and Zr in each of the effluent streams and the overall material balance for each component are given in Tables 5 and 6. It should be noted that the percentages of components in the solvent are not included in the overall material balance since the solvent was recycled during testing. Distribution coefficients were calculated for total alpha, ^{137}Cs, ^{90}Sr, ^{241}Am, ^{238}Pu, ^{239}Pu, and ^{154}Eu on various stages. The resulting distribution coefficients are given in Table 7. A discussion of the behavior of each component follows.

Cesium. The ^{137}Cs activity was reduced from 3.24E+06 dps/mL in the feed to 1.44E+04 dps/mL (0.39 Ci/m^3) in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 99.4%. This somewhat low ^{137}Cs removal efficiency was due to low distribution coefficients which ranged from 0.68 to 1.3 in the extraction section. In the strip section distribution coefficients ranged from 0.11 to 0.20, resulting in 99.99% of the extracted Cs exiting in the strip product.

Although the activity of ^{137}Cs was reduced to 0.39 Ci/m^3 which is below the NRC Class A LLW limit of 1.0 Ci/m^3, the final activity in a grout waste form is dependent upon the waste loading obtained. It is expected that a removal efficiency of approximately 99.9% or greater would be required to assure NRC Class A LLW limits are met in the grout. The addition of more extraction stages would ensure this result.

Table 6. Percentages of metals in the effluent streams for the flowsheet demonstration with WM-185 waste.

Effluent	Al	Ba	Ca	Ce	Fe	Hg
Raffinate	108.2%	<1.05%	97.8%	<11.4%	74.7%	110.9%
Strip	0.14%	105.7%	9.9%	109%	8.3%	<1.2%
Wash	---	---	---	---	---	---
Solvent	0.022%	3.9%	<1.2	<25.8%	0.44%	0.04%
Material Balance	108.4%	106.8%	107.7%	109% - 120%	83.0%	110.9%
Effluent	K	Mo	Na	Nd	Pb	Zr
Raffinate	74.6%	79.1%	108.0%	<2.4%	1.2%	13.0%
Strip	27.9%	31.7%	0.13%	112%	83.2%	68.6%
Wash	0.008%	---	0.01%	---	---	---
Solvent	0.15%	<15.6%	0.02%	<5.5%	<0.62%	0.09%
Material Balance	102.4%	110.8%	109.0%	112% - 114.4%	84.4%	81.6%

Table 7. Distribution coefficients from the flowsheet demonstration with WM-185 tank waste.

Stage		D_{Cs-137}	D_{Sr-90}	D_{Eu-154}	Total alpha	D_{Am-241}	D_{Pu-238}	D_{Pu-239}
Extraction	24	0.77	2.7	---	8.7	22.4	5.7	14.7
	23	0.73	1.6	---	13.4	14.7	4.1	0.8
	22	0.83	5.6	---	10.9	---	---	---
	21	0.78	0.69	---	6.2	11.2	7.2	25.2
	20	0.90	2.9	---	11.6	---	---	---
	19	1.0	3.3	---	7.2	6.4	6.6	18.6
	18	1.1	2.5	---	12.2	---	---	---
	17	1.33	2.6	>1.7	27.7	9.8	191	255
	16	0.68	6.3	>2.8	66.8	---	---	---
	15	0.77	2.2	>7.2	92.0	6.6	308	326
Scrub	14	31.1	537	---	1920	---	---	---
	13	18.7	692	>197	1754	---	---	---
Strip	12	0.11	2.9E-05	<0.01	7.4E-04	2.8E-04	2.6E-04	2.5E-04
	11	0.15	4.4E-04	<0.01	0.012	0.006	0.007	0.008
	10	0.20	0.047	<0.005	0.012	---	---	---
	9	0.19	---	<0.007	---	0.001	0.002	0.010
	8	0.19	0.09	<0.04	0.07	---	---	---
	7	0.19	0.06	---	0.54	0.005	0.005	0.007
Wash	6	0.84	---	>3.8	20.1	---	---	---
	5	1.2	---	>3.2	126	---	---	---
	4	1.9	---	>3.2	134	---	---	---
	3	1.5	---	>1.5	196	---	---	---
	2	2.2	---	>2.4	56.2	---	---	---
	1	3.0	---	>2.2	21.8	---	---	---

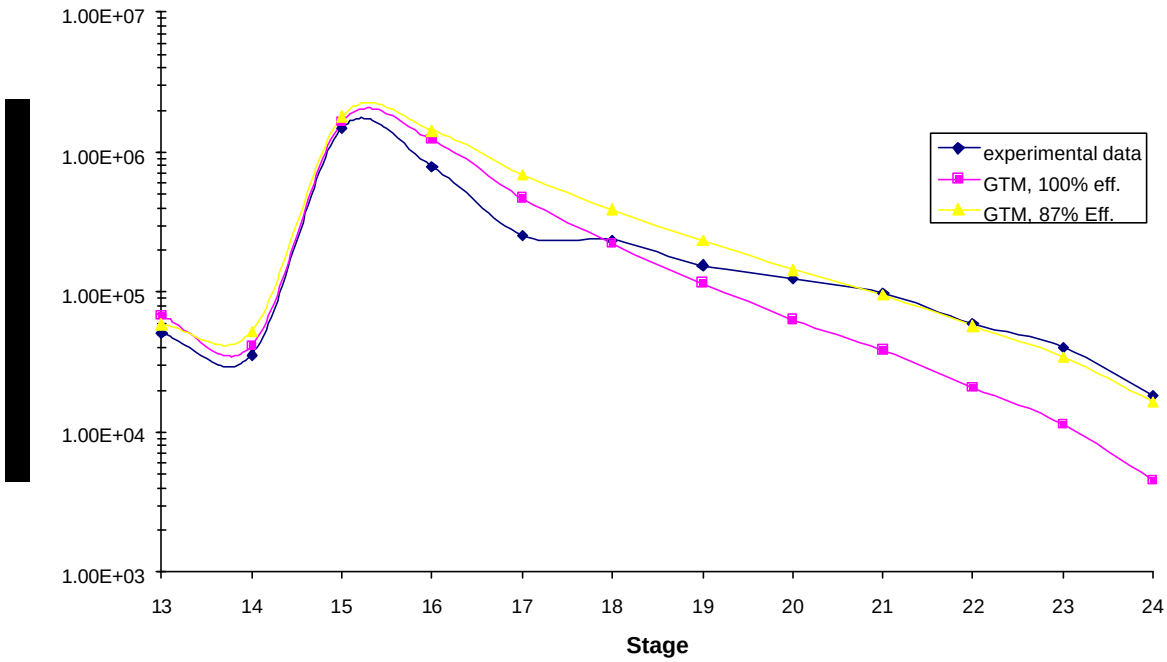

Figure 4. Comparison of aqueous phase ^{137}Cs experimental data with GTM data at 87% and 100% efficiency in the extraction section.

The ^{137}Cs distribution coefficients in the extraction section were used in conjunction with the Generic TRUEX Model (GTM) to determine the approximate stage efficiency at which the centrifugal contactors were operating.[12] As shown in Figure 4, an efficiency of 87% results in a predicted raffinate activity for ^{137}Cs which is comparable to the experimental results. If the contactors were operating at an efficiency of 100%, the activity of the raffinate is expected to approach 4.55E+03 dps/mL (0.12 Ci/m^3). It is expected that an efficiency greater than 95% would be obtained using full scale centrifugal contactors which are designed specifically for the UNEX process.

The raffinate from the test was collected from the time the WM-185 feed was introduced into the contactors until shutdown. A composite sample of this solution was analyzed for ^{137}Cs and an activity of 4.66E+03 dps/mL was obtained. This result confirms the analytical results from samples taken during the run which show that the ^{137}Cs activity in the raffinate remained low throughout the run.

Strontium. The ^{90}Sr activity was reduced from 2.76E+06 dps/mL in the feed to 111 dps/mL (0.003 Ci/m^3) in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 99.995% which is sufficient to reduce the ^{90}Sr activity of the WM-185 waste below the NRC Class A LLW criteria of 0.04 Ci/m^3. The final activity of ^{90}Sr in a grout waste form is dependent upon the waste loading obtained but is expected to be below the Class A limit with a 99.995% removal efficiency.

Distribution coefficients for ^{90}Sr ranged from 0.7 to 6.3 in the extraction section with an average value of approximately 3.0. In the strip section distribution coefficients for ^{90}Sr ranged from 2.9E-05 to 0.09. As a result, 99.9999% of the extracted ^{90}Sr exited in the strip product.

The raffinate from the test was collected from the time the WM-185 feed was introduced into the contactors until shutdown. A composite sample of this solution was analyzed for ^{90}Sr and an activity of

22.6 dps/mL was obtained. This result confirms the analytical results from samples taken during the run which show that the ^{90}Sr activity in the raffinate remained low throughout the run.

Actinides. The total alpha activity was reduced from 2.33E+04 dps/mL in the feed to 7.2 dps/mL (0.16 nCi/g) in the aqueous raffinate immediately prior to shutdown. This corresponds to a removal efficiency of 99.96% which is sufficient to reduce the actinide activity of the WM-185 waste below the NRC Class A non-TRU criteria of 10 nCi/g. Removal efficiencies of 99.999% and 99.994% were obtained for ^{241}Am and ^{238}Pu, respectively. In the strip section, 99.97%, 99.996%, and 99.9999% of the total alpha, ^{241}Am, and ^{238}Pu, respectively, were removed from the solvent.

The removal efficiency for total alpha was greatly improved as compared to previous flowsheet testing. It the previous testing, only 95% removal was obtained due to loading of the Ph_2Bu_2CMPO with Zr, Fe, and Mo. The following changes were made to the previous flowsheet, which reduced the loading of Ph_2Bu_2CMPO during this testing.

- An increased amount of fluoride was added to the feed solution to complex the Zr and Fe.
- The feed solution was diluted, thus decreasing the concentration of Zr, Fe, and Mo.
- A scrub solution consisting of 0.3 *M* citric acid was used instead of 0.1 *M* HF.

In the previous flowsheet testing it is estimated that nearly all of the Ph_2Bu_2CMPO was consumed from the extraction of Zr, Fe, and Mo. It is estimated that 52% of the Ph_2Bu_2CMPO was consumed with this modified flowsheet based on the assumption of three moles of Ph_2Bu_2CMPO consumed per mole of the extracted metal.

The raffinate from the test was collected from the time the WM-185 feed was introduced into the contactors until shutdown. A composite sample of this solution was analyzed for total alpha and an activity of 8.3 dps/mL was obtained. This result confirms the analytical results from samples taken during the run which show that the actinide activity in the raffinate remained low throughout the run.

Technetium. Less than 0.14% of the ^{99}Tc was extracted from the WM-185 waste by the universal solvent and exited in the strip product. The overall material balance for ^{99}Tc was 81.3%. The wash effluent was not analyzed for ^{99}Tc; therefore, any ^{99}Tc in the wash effluent would increase the overall material balance. It is important to note that the ^{99}Tc activities in INTEC tank wastes are anticipated to be below NRC Class A LLW requirements (0.3 Ci/m^3). Technetium removal is of concern due to its mobility, as TcO_4^-, in the environment. It would therefore be advantageous to be able to fractionate ^{99}Tc from the wastes.

Aluminum, barium, calcium, cerium, iron, mercury, potassium, molybdenum, sodium, neodymium, lead, and zirconium. The effluent streams were analyzed for Al, Ba, Ca, Ce, Fe, Hg, K, Mo, Na, Nd, Pb, and Zr. Of these components, Ba, Ce, Nd, and Pb were nearly completely extracted. Significant amounts of Zr, Mo, K, and Fe were also extracted (87%, 32%, 28%, and 8%, respectively). The strip product analytical results indicate that 10% of the Ca was extracted and exited in the strip product. However, the raffinate analytical results indicate that 98% of the Ca remained in the raffinate. Aluminum, mercury, and sodium were essentially inextractable (<1% extracted).

Approach to Steady State

The activities of ^{137}Cs, ^{90}Sr, and total alpha as a function of time (T_0 = start of WM-185 feed) are given in Figures 5 and 6 for the aqueous raffinate and strip product, respectively. It is assumed that steady state was reached when the activity of the components varied by less than the analytical error associated with the sample analyses.

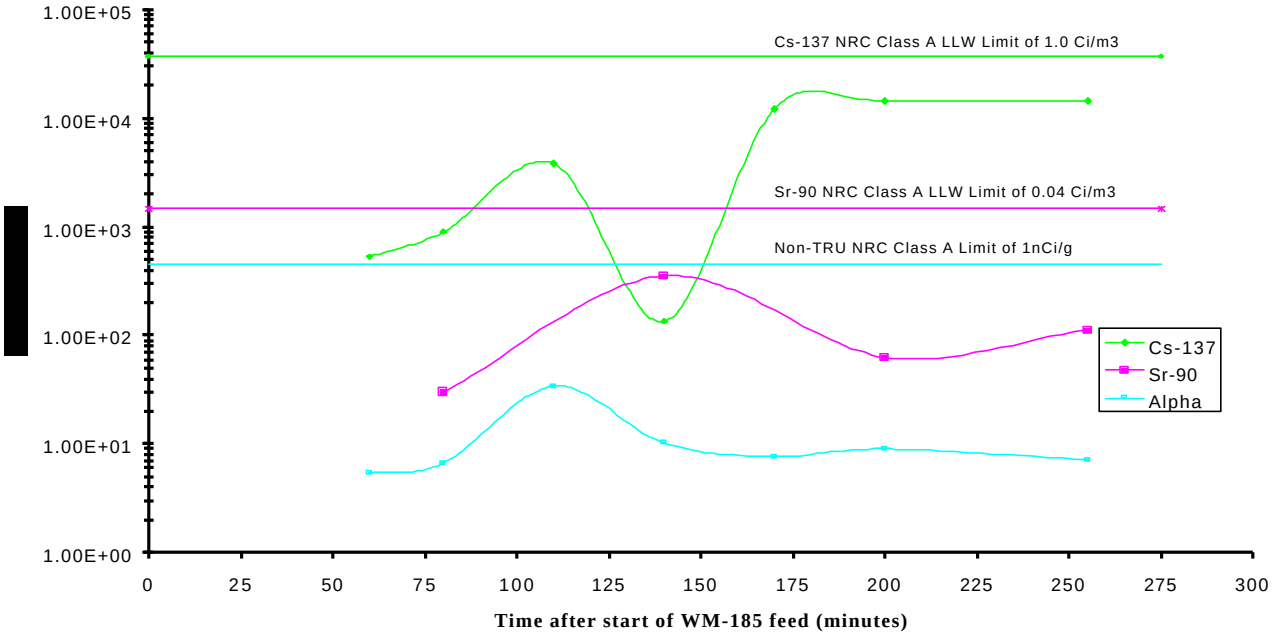

Figure 5. Approach to steady state for total alpha, ^{137}Cs, and ^{90}Sr in the raffinate.

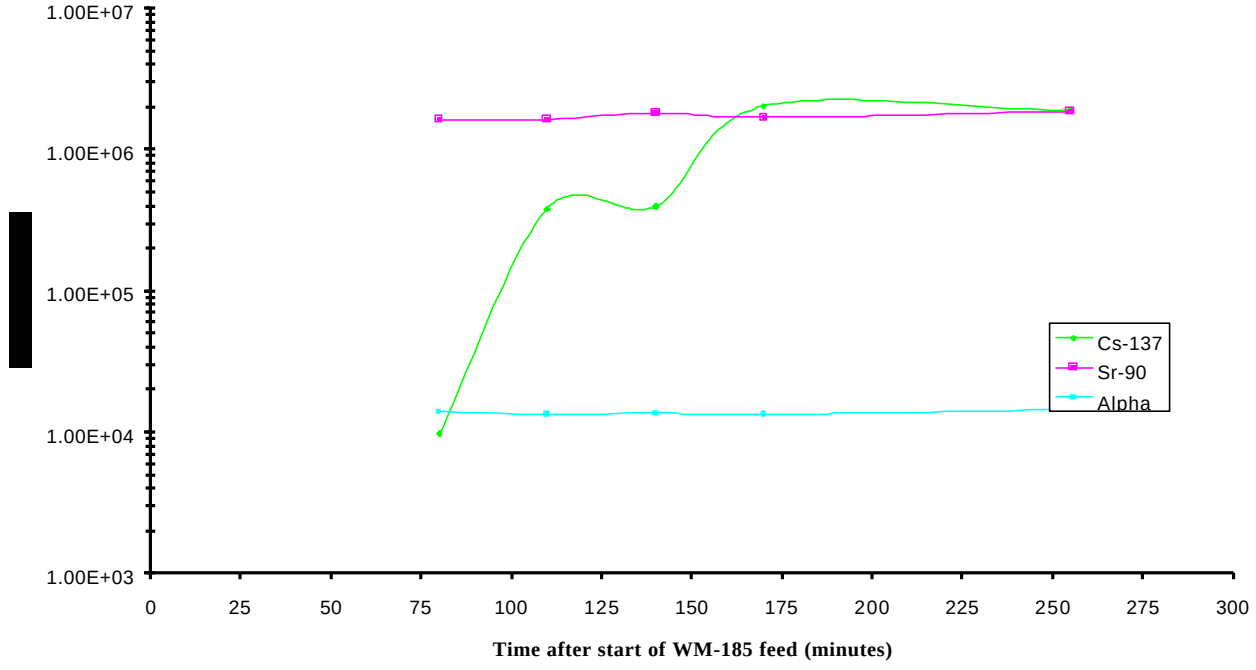

Figure 6. Approach to steady state for total alpha, ^{137}Cs, and ^{90}Sr in the strip product.

The approach to steady state in the aqueous raffinate was evaluated for ^{137}Cs, ^{90}Sr, and total alpha. Steady state was reached within 170 minutes for ^{137}Cs. The ^{137}Cs activity remained below the NRC Class A LLW limit of 1.0 Ci/m^3 throughout the test. The approach to steady state can not be evaluated for ^{90}Sr in the aqueous raffinate since the analytical results were very close to detection limits and have large uncertainties associated with them (>50%). All analyses were below the NRC Class A LLW criteria of 0.04 Ci/m^3 even with the large uncertainties factored in. A sample of the composite raffinate at the end of the test was analyzed for ^{90}Sr and an activity of 22.6 dps/mL ±36% (0.0006 Ci/m^3) was obtained. Steady state was reached for the total alpha activity within 140 minutes of the start of WM-185 feed. The activity of total alpha remained well below the NRC Class A non-TRU limit of 10 nCi/g throughout the test.

The approach to steady state in the strip product was evaluated for ^{137}Cs, ^{90}Sr, and total alpha. Steady state was reached for ^{90}Sr and total alpha within 80 minutes from the start of WM-185 feed. Steady state was reached within 170 minutes for ^{137}Cs. With the ^{137}Cs distribution coefficients in the extraction section less than one, it is expected to take longer to reach steady state than for ^{90}Sr and total alpha with distribution coefficients well above one.

CONCLUSIONS AND RECOMMENDATIONS

Conclusions

The universal solvent extraction process, developed as a joint effort between the INEEL and the Khlopin Radium Institute, is a viable process for the separation of Cs, Sr, and the actinides from INTEC SBW. Overall removal efficiencies of 99.4%, 99.995%, and 99.96% were obtained for ^{137}Cs, ^{90}Sr, and total alpha, respectively, with the flowsheet demonstration using WM-185 waste. This is sufficient to reduce the activities of ^{137}Cs, ^{90}Sr, and the actinides in the raffinate to below NRC Class A LLW limits. Because this testing was performed in a limited number of small-scale centrifugal contactors, which were not designed for specifically for this process, it is expected that higher removal efficiencies could easily be achieved through contactor design and scale-up, and optimization of the number of stages.

The actinides, ^{137}Cs, and ^{90}Sr were effectively removed from the universal solvent with the six stages of 1 *M* guanidine carbonate, 20 g/L DTPA strip in the universal solvent flowsheet demonstration.

The effluent streams from the flowsheet demonstration were also analyzed for ^{99}Tc, Al, Ba, Ca, Ce, Fe, Hg, K, Mo, Na, Nd, Pb, and Zr. Ba, Ce, Nd, and Pb were nearly completely extracted. Significant amounts of Zr, Mo, K, Ca, and Fe were also extracted (87%, 32%, 28%, 10%, and 8%, respectively). Technetium, aluminum, mercury and sodium were essentially inextractable (<1% extracted).

Based on the analytical results for ^{137}Cs and computer modeling with the Generic TRUEX Model, the centrifugal contactors were operating at a stage efficiency of approximately 87% in the extraction section.

The positive results obtained from this flowsheet test have resulted in the initiation of a Feasibility Study of the UNEX process for the treatment of INTEC SBW by an architect-engineering company. This feasibility study will be performed in FY 2000 and will define facility and equipment requirements and generate a cost estimate.

Recommendations

Further development and testing of the universal solvent extraction process is recommended in order to optimize the flowsheet and support the Feasibility Study. Adjustments to the flowsheet should be made to minimize the flowrates of process solutions. Specifically, reduction of the O/A ratio in the extraction section, reduction of the strip flowrate and/or guanidine carbonate/DTPA concentration, and minimization of the wash effluent flow via recycle should be investigated. Changes in these areas would decrease the overall amounts of high and low-activity liquid wastes generated by the process and potentially reduce the quantity of high and low-activity waste forms generated. Additionally, alternative scrub solutions should be developed in the event that the citrate concentration in the low-activity waste is determined to be unacceptable for the formation of a grout waste form.

Long term testing of the UNEX process with simulated or actual waste solution is recommended. This type of testing will determine whether any metals buildup in the solvent and adversely affect the extraction of the radionuclides. Also, any reduction in the performance of the solvent due to solvent degradation or extractant losses can be evaluated.

Finally, the development of the UNEX process for the treatment of dissolved INTEC calcine should continue. Initially, testing with dissolved calcine surrogates should be performed to develop a flowsheet for future countercurrent tests.

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Appendix A: Experimental Data

Table A-1. UNEX process flowsheet demonstration feed and effluent activities.

Sample	Sr-90 *dps/mL*	Cs-137 *dps/mL*	Eu-154 *dps/mL*	Gross Alpha *dps/mL*	Am-241 *dps/mL*	Pu-238 *dps/mL*	Pu-239 *dps/mL*	Tc-99 *dps/mL*
Raff. Rinse		269	<1.14	5.53E-03				
Strip Rinse		201	<1.64	11.5				
Wash Rinse		45.4	<1.19	3.48E-02				
WM-185 Feed	2.76E+06	3.24E+06	8.82E+03	2.33E+04	2.20E+03	1.93E+04	2.36E+03	1.14E+03
Aq. Raff - 1	0	532	<97.30	5.41				
Aq Raff - 2	30	896	<137	6.7	0.15	1.13	0.31	96.8
Strip - 2	1.63E+06	9.76E+03	24.7	1.40E+04	1.26E+03	1.14E+04	1.27E+03	<1.0
Wash - 2	2.28	144	<1.09	9.78E-03				
Org - 2	0.684	40.5	<1.90	0.945				
Aq Raff - 3	0	3.82E+03	<53	34.0				
Strip - 3	1.63E+06	3.80E+05	1.18E+03	1.34E+04				
Wash - 3	7.35	196	<1.47	0.175				
Org - 3	2.6	520	2.66	2.1				
Aq Raff - 4	352	134	<1.43	10.23				
Strip - 4	1.81E+06	3.95E+05	1.09E+03	1.36E+04				
Wash - 4	3.67	143	<0.86	6.32E-03				
Org - 4	6.8	203	<2.39	2.33				
Aq Raff - 5	0	1.21E+04	<44.2	7.65				
Strip - 5	1.68E+06	2.02E+06	3.57E+03	1.33E+04				
Wash - 5	1.35	138	<1.32	0.135				
Org - 5	13	48.6	1.13					
Aq Raff - 6	63	1.44E+04	<29.2	9				
Raff - Final	111	1.44E+04	28.6	7.2	3.90E-03	0.95	0.043	715.8
Strip - Final	1.85E+06	1.89E+06	4.30E+03	1.45E+04	1.44E+03	1.16E+04	1.52E+03	<1.000
Wash - Final	4.5	117	<0.82	0.13				
Org - Final	16	68	2.6	1.87	2.32	0.35	5.30E-03	0.06
Composite samples								
Aq Raff - 10	22.6	4.66E+03	<43.4	8.26				
Strip - 10	1.75E+06	3.69E+05	1.12E+03	1.28E+04				
Wash - 10	0.56	125	<1.28	0.028				
Org - 10	1.4	900	<137	1.2				

Table A-2. Stable metals analysis data from the UNEX flowsheet demonstration.

Sample	K *mg/L*	Pb *mg/L*	Hg *mg/L*	Zr *mg/L*	Ba *mg/L*	Ca *mg/L*	Fe *mg/L*	Na *mg/L*	Al *mg/L*	Mo *mg/L*	Ce *mg/L*	Nd *mg/L*
Strip Blank								7.08				
WM-185 Feed	5.33E+03	368.7	655.2	950.1	6.76	1.99E+03	1.20E+03	2.44E+04	1.38E+04	21.2	6.2	9.4
Aq Raff - 2	1.18E+04	2.84	1067.2	35.8	<0.055	1.37E+03	625.3	1.78E+04	1.10E+04	12.5	<0.55	<0.18
Strip - 2	1.02E+03	1.56	<1.0	1.06	0.055	16.6	0.073	463	0.57	0.61	0.175	0.55
Aq Raff - Final	3.08E+03	3.42	563.1	95.6	<0.055	1.51E+03	696.2	1.84E+04	1.15E+04	13.0	<0.55	<0.175
Strip - Final	921	190.2	<5.0	404.0	4.43	122.1	61.6	402	11.9	4.17	4.18	6.52
Wash - Final	0.26							13.3				
Org - Final	3.17	<0.90	0.108	0.35	0.10	<9.56	2.09	444	1.18	<1.30	<0.63	<0.20

Table A-3. Stage activities from UNEX flowsheet demonstration.

	Sample	Sr-90 *dps/mL*	Cs-137 *dps/mL*	Eu-154 *dps/mL*	Alpha *dps/mL*	Am-241 *dps/mL*	Pu-238 *dps/mL*	Pu-239 *dps/mL*
wash	St1-O		30.8	2.94	1.85			
	St1-A		10.2	<1.32	8.48E-02			
	St2-O		62.2	2.22	1.4			
	St2-A		28.8	<0.93	2.49E-02			
	St3-O		37.7	1.83	4.11			
	St3-A		25.9	<1.19	2.10E-02			
	St4-O		47.8	3.2	2.72			
	St4-A		25.2	<0.99	2.03E-02			
	St5-O		56.6	3.44	3.52			
	St5-A		49.2	<1.09	2.79E-02			
	St6-O	8.8	72.8	3.99	3.02			
	St6-A		86.3	<1.27	0.15			
strip	St7-O	0.90	133	<0.86	2.06	0.038	0.014	4.10E-03
	St7-A	15.5	702	<3.77	3.8	8.32	2.64	0.55
	St8-O	1.7	595	<1.04	0.854			
	St8-A	18.21	3162	23.2	11.7			
	St9-O	0	3.76E+03	<1.40	0	0.059	3.40E-02	2.30E-02
	St9-A	40.4	1.99E+04	192	53.91	92.6	14.7	2.31
	St10-O	5.1	1.74E+04	<3.35	2.47			
	St10-A	108	8.68E+04	644	210			
	St11-O	1.8	6.74E+04	<11.90	8.26	3.45	2.56	0.355
	St11-A	4.13E+03	4.66E+05	1286	676	536	359	47.2
	St12-O	39.4	1.78E+05	<35.70	6.33	0.473	2.38	0.286
	St12-A	1.38E+06	1.60E+06	5.73E+03	8.60E+03	1.66E+03	9.30E+03	1.16E+03
scrub	St13-O	8.65E+05	9.54E+05	2.09E+03	6.91E+03			
	St13-A	1.25E+03	5.10E+04	<10.60	3.94			
	St14-O	9.12E+05	1.11E+06	<938.0	7.24E+03			
	St14-A	1.70E+03	3.57E+04	<7.45	3.77			
extraction	St15-O	1.25E+06	1.15E+06	4.70E+03	1.26E+04	1.08E+03	1.02E+04	1.24E+03
	St15-A	5.78E+05	1.49E+06	<648	1.37E+02	164	33.1	3.8
	St16-O	6.68E+05	5.36E+05	1.02E+03	1.67E+03			
	St16-A	1.06E+05	7.90E+05	<365.8	2.50E+01			
	St17-O	8.25E+04	3.37E+05	227	302	62.4	193	23.7
	St17-A	3.19E+04	2.53E+05	<130	10.9	6.35	1.01	0.093
	St18-O	1.88E+04	2.51E+05	<139	65.7			
	St18-A	7.52E+03	2.23E+05	<80.1	5.4			
	St19-O	4.82E+03	1.57E+05	<30.5	25.3	3.81	7.61	1.77
	St19-A	1.45E+03	1.55E+05	<42.3	3.51	0.6	1.15	0.095
	St20-O	1.14E+03	1.11E+05	<20.6	16.1			
	St20-A	393	1.24E+05	<31.8	1.39			
	St21-O	183	7.71E+04	<14.2	18.9	2.28	6.08	1.56
	St21-A	266	9.89E+04	<24.4	3.04	0.203	0.845	0.062
	St22-O	462	4.94E+04	<9.2	18.8			
	St22-A	82.7	5.97E+04	<14.8	1.72			
	St23-O	42.4	2.94E+04	<5.87	18.0	1.88	4.68	1.64
	St23-A	27.2	4.03E+04	<11.4	1.34	0.128	0.554	0.047
	St24-O	67.5	1.40E+04	<4.13	13.6	2.46	4.08	0.811
	St24-A	24.7	1.82E+04	<7.81	1.57	0.11	0.72	0.055

Table A-4. Batch contact data with WM-185.

Sample	Cs-137 *(dps/mL)*	Eu-154 *(dps/mL)*	Sr-85 *(dps/mL)*	Sr-90 *(dps/mL)*
Feed-1	3.01E+06	7.20E+03	2.95E+04	2.48E+06
Feed 1 = 154mL WM-185 + 15.4mL 5.2 M HF + 30.6 mL 0.3M GC/0.05M HNO_3 + 0.55 mL Sr-85				
E1-O	1.66E+06	6.61E+03	2.11E+04	---
E1-A	1.42E+06	<6.03E+02	7.79E+03	7.49E+05
E1-OS[1]	2.97E+05	1.20E+03	3.87E+03	1.65E+06
Dist.	1.17	10.96	2.71	---
Dist. (Strip)	0.21	1.99	0.50	2.20
%MB	102.3	100.2	97.9	---
%MB (Strip)	57.0	25.0	39.5	96.7
E2-O	2.37E+06	1.15E+04	3.16E+04	---
E2-A	2.23E+06	2.06E+03	1.76E+04	---
Dist.	1.06	5.58	1.80	---
%MB	98.5	98.2	97.2	---
E3-O	2.84E+06	1.56E+04	3.80E+04	---
E3-A	2.63E+06	4.09E+03	2.42E+04	---
Dist.	1.08	3.81	1.57	---
%MB	101.7	105.3	101.8	---
E4-O	3.02E+06	1.74E+04	3.66E+04	---
E4-A	2.86E+06	4.92E+03	2.87E+04	2.66E+06
E4-OS	2.96E+06	1.16E+04	3.68E+04	3.54E+06
Dist.	1.06	3.54	1.28	---
Dist. (Strip)	1.03	2.36	1.28	1.33
%MB	100.51	97.9	96.74	---
%MB (Strip)	99.5	72.46	97.0	---
ST1-O	4.26E+05	7.76E+03	<4.63E+02	---
ST1-A	2.42E+06	7.81E+03	3.42E+04	---
Dist.	0.18	0.99	<0.01	---
%MB	---	---	---	---
ST2-O	7.06E+04	2.14E+03	<2.05E+02	---
ST-2A	3.34E+05	4.84E+03	<4.01E+02	---
Dist.	0.21	0.44	---	---
%MB	95.0	90.0	---	---
Feed-2	4.00E+06	7.72E+03	---	3.49E+06
Feed 2 was unadjusted WM-185 w/o Sr-85				
E5-O	1.54E+06	1.32E+03	---	---
E5-A	2.28E+06	6.53E+03	---	1.23E+06
E5-OS	1.51E+06	2.89E+03	---	2.11E+06
Dist.	0.68	0.20	---	
Dist. (Strip)	0.66	0.44	---	1.72
%MB	95.50	101.68	---	
%MB (Strip)	94.75	122.02	---	95.70

[1]EX-OS samples were stripped with 1.5M guanadine carbonate (GC)/10 g/L DTPA